Final Technical Report

Reactive Multiphase Behavior of CO_2 in Saline Aquifers Beneath the Colorado Plateau

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ABSTRACT

Over 100 million tonnes (Mt) of CO_2 is released to the atmosphere each year from power plants located in the Colorado Plateau and southern Rocky Mountains region of the U.S. This region contains broad anticlinal structures, not generally thought of as gas traps, that may be suitable for permanent sequestration of CO_2. Field studies and numerical simulations of naturally occurring CO_2 reservoirs at Springerville-St. Johns, AZ-NM, Farnham Dome, UT and Crystal Geysers-Ten Mile Graben, UT have been conducted to quantitatively evaluate the factors that control the mobility of CO_2 under geologic conditions. Soil gas flux measurements show no evidence of leakage today at Springerville-St. Johns and at Farnham Dome, although there is clear evidence that CO_2 was discharged to the atmosphere in the past. At Crystal Geysers-Ten Mile Graben, localized points of CO_2 discharge occur along fault zones. The bulk of the CO_2 at these sites is stored as gas accumulations and as dissolved species in the ground water. These observations suggest that monitoring the leakage of CO_2 will present major challenges.

The numerical simulations suggest that the anticlimal structures could serve as CO2 repositories. The models suggest approximately 70% of the injected CO_2 can be permanently sequestered. The simulations predict that minerals may trap up to 20% of the CO_2 but investigations of core samples from Springerville-St. Johns suggest the sequestration of CO_2 in mineral phases in this field has been insignificant and in general, may be less important than previously considered. The importance of mineral trapping requires additional attention.

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EXECUTIVE SUMMARY

Few naturally occurring CO_2 reservoirs have been investigated as analogues of sequestration sites. The Colorado Plateau-southern Rocky Mountain region of the US contains natural CO_2 reservoirs, geologic structures that may be suitable for permanent sequestration of CO_2 and collocated power plants that discharge 100 million tonnes (Mt) of CO_2 to the atmosphere each year. Field studies were conducted at three sites within the region: Springerville-St. Johns, Arizona-New Mexico, Farnham Dome, Utah and Crystal Geysers-Ten Mile Graben, Utah. Field observations and numerical simulations were utilized to address a number of specific sequestration issues. These included the sequestration of CO_2 by trapping under low permeability seals, direct precipitation of CO_2-bearing minerals through reactions with saline aquifer fluids, fluid-mineral reactions leading to the precipitation of new minerals, fluid-fluid reactions and their significance in controlling. The investigations conclude that broad anticlinal structures, not generally thought of as gas traps, may be suitable for permanent sequestration of CO_2 and that the CO_2 will occur principally as gas accumulations and as dissolved carbonate species in the ground water. Outflows of these waters represent a continuous source of leakage of CO_2 from the reservoirs, and present particular challenges for long term monitoring. Leakage of CO_2 to the atmosphere was observed at isolated locations along faults zones at Crystal Geysers-Ten Mile Graben but not at the other sites, although geologic evidence of past discharges has been found. The locations of these points of leakage cannot be predicted. Permanent trapping of CO_2 in mineral phases was found to be insignificant at Springerville-St. Johns, but the numerical simulations suggest it may be important elsewhere in the region. Additional field studies are needed to assess the significance of this trapping mechanism.

EXPERIMENTAL

Not applicable.

RESULTS AND DISCUSSION

Background

The production of over 10,000 MW of electricity from power plants located within the Colorado Plateau and adjacent Rocky Mountain region results in the emission of over 100 million tonnes (Mt) of CO_2 each year to the atmosphere. This region contains numerous natural CO_2 fields (Fig. 1). Several of these fields are presently in production, with most of the CO_2 (25 Mt/year) being piped 800 km to enhanced oil recovery projects in west Texas.

The occurrence of natural CO_2 reservoirs around the Colorado Plateau, widespread saline aquifers at depth, and bicarbonate aquifers at shallow depth offer the potential to sequester all the CO_2 currently being emitted from the local coal-fired power plants. At least three large natural CO_2 reservoirs have been significantly depleted due to past production (Farnham Dome, UT; McElmo Dome, CO; and Bravo Dome, NM), and have proven storage potential available. Undeveloped reservoirs (e.g. Springerville, AZ, Gordon Creek, UT) and analogous geologic structures that do not contain CO_2 may also serve as repositories for additional CO_2. When these factors are combined with the existing CO_2 pipelines linking the region with west Texas, the Colorado Plateau would appear to be strategically important in the U.S. for demonstrating the feasibility of long-term subsurface CO_2 sequestration.

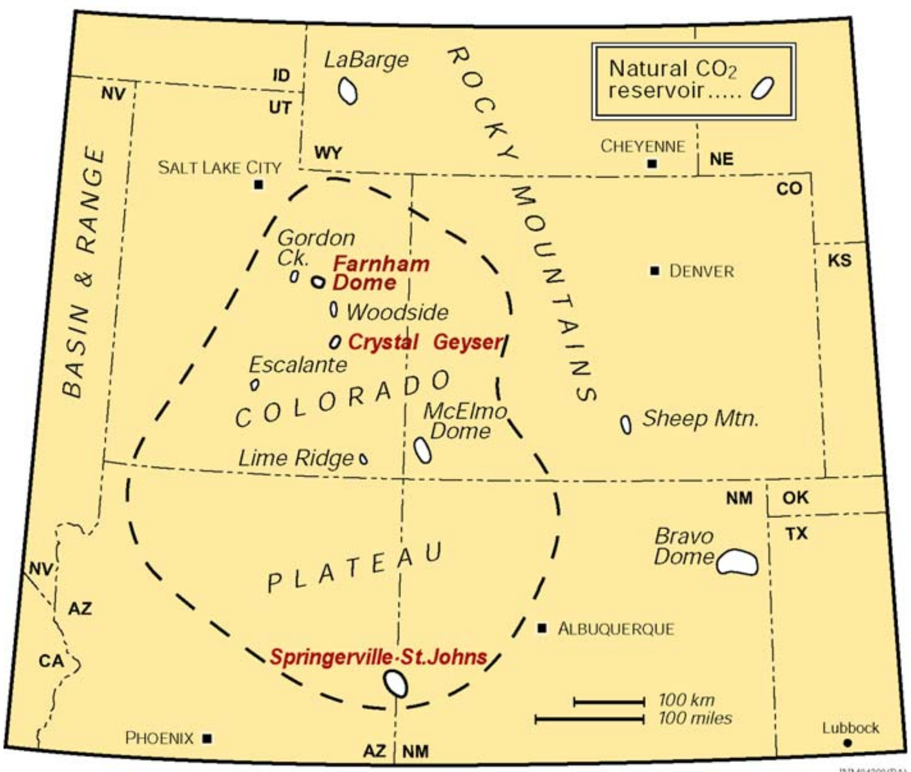


Fig. 1. Natural CO_2 occurrences on the Colorado Plateau, with the three occurrences discussed in this paper highlighted in red. Crystal Geyser is the site of CO_2 seepages along the Little Grand Wash fault zone, and in the Ten Mile Graben.

The purpose of the research conducted under this investigation was to use the CO_2 reservoirs that occur beneath the Colorado Plateau as natural analogues for quantitatively evaluating the effects of CO_2 sequestration in the U.S. Rocky Mountain – Colorado Plateau region. A variety of sequestration strategies and environmental issues were explicitly considered. These included the sequestration of CO_2 by trapping under low permeability seals, direct precipitation of CO_2-bearing minerals through reactions with saline aquifer fluids, fluid-mineral reactions leading to the precipitation of new minerals, fluid-fluid reactions and their significance in controlling the mobility of CO_2 and the escape of CO_2 to the atmosphere.

Previous Investigations

Prior to the initiation of our investigation, direct study of the reactive effects of CO_2 on the Colorado Plateau reservoir rocks appears to be limited to the unpublished Bravo Dome report of Amoco (Nelis, 1994), referred to by Pearce et al., (1996). Fouret (1982, 1996) investigated the depositional history and diagenesis at the Lisbon field, where late-stage CO_2 should have left an overprint. A request by the PI (Allis) to BP-Amoco for release of the Nelis report resulted in a deferral for the moment while this company establishes its CO_2 research program (S. Baines, D. Thomas, email comms., 12/99 appended in A.3). The communications left open the possibility of collaboration next year. However the key results from Nelis (1994) are described in Pearce et al. (1996). Feldspars are corroded, but some of this is attributed to early phases of burial. Ferroan dolomite, kaolinite, zeolites, and in one area of the field, chlorite, occur as late overgrowths or infilling minerals. There is also evidence of removal of earlier kaolinite. Zeolite formation appears strongest along the northwest-trending fault/flexure of the reservoir. Zones of high porosity graded into zones of much reduced porosity due to the presence of authigenic cements such as gibbsite and kaolinite.

At the Lisbon field (UT), the carbonate reservoir is characterized by moldic porosity, where calcitic fossils have been leached from their dolomite host. However the overlying limestone "packstone" has lower porosity and permeability, and not been leached (Fouret, 1996). Late stage silicification, and pore-filling secondary anhydrite, dolomite and calcite are also present.

Elsewhere on the Colorado Plateau, bleaching of red sandstone aquifers has been attributed to reduction of hematite due to acidic fluids (Sanford, 1995). The cause of the acidity was attributed to organic acids associated with kerogen maturation, but we believe migration of CO_2 gas is more likely (e.g. see Killops et al., 1996; CO_2 is related to early maturation of coal, immediately preceding oil generation). Sanford (1995) points out that immediately prior to oil migration in the White Rim Sandstone around the Green and Colorado Rivers, dissolution of early calcite cement, silica overgrowths and kaolinite formation occurred as a result of acidic pore fluid interactions. At a nearby site, Breit and Goldhaber (1996) suggest that late stage reducing solutions, perhaps as saline fluids rising up fault zones, have caused precipitation of calcite, dolomite and barite in the overlying aquifers. Similarly, close to the Moab fault (east Utah), late-stage reducing solutions have caused calcite, ankerite, barite and pyrite cements and veining (Foxford et al., 1996). These solutions may be important analogues for assessing and simulating the effects of leaking CO_2-rich fluids from CO_2 sequestration projects.

Project Results

Field investigations and numerical simulations were conducted at Springerville-St. Johns (AZ-NM) and in Utah at Farnham Dome, Crystal Geyser and Ten Mile Graben. The results have been presented at the 1st, 2nd, 3rd, and 4th National Conferences on Carbon Sequestration, the 6th International Conference on Greenhouse Gas Control Technologies, held in Kyoto, Japan, at the annual meetings of the Geological Society of America, American Geophysical Union and Association of Petroleum Geologists and in a special volume of Chemical Geology on "Geochemical Aspects of CO_2 Sequestration". A list of the papers generated as a result of this project is presented in the References. The full texts of the papers presented in Chemical Geology are included as Appendix 1 and 2.

Springerville-St. Johns, Arizona-New Mexico

Springerville-St. Johns provides a natural example of a leaky reservoir (Fig. 2; Appendix 1). The CO2 occurs in Permian siliciclastic rocks of the Supai Formation at depths of less than ~800 m. Shales and anhydrite beds form low permeability seals. At the surface, extensive travertine (calcium carbonate) deposits record the effects of CO2 discharged to the atmosphere by spring waters. Much of this CO2 may have been derived from the dissolution of limestone within the reservoir. 14C AMS dating of pollen trapped in the travertine deposits suggest that the system began to leak at ~3500 years BP (Table 1, Fig. 2). The youngest dated deposit had an age of 887 years BP. The maximum difference in elevation between the lowest and highest dated sample is 338 m. Although springs are associated with many of the deposits, there is no evidence of travertine precipitation today.

Faults that cut the underlying preCambrian granite serve as conduits for the upward migration of the gas. Flux measurements indicate that CO2 no longer reaches the surface. However, chemical analyses of ground waters record high concentrations of bicarbonate, documenting the continued flux of CO2 from depth. These fluids migrate down the hydrologic gradient, dispersing the CO2 away from the reservoir. The ultimate fate of the CO2 transported in ground waters is not known. Investigations of rock samples taken from cores drilled into the reservoir indicate that little CO2 has been permanently sequestered in mineral phases. Only traces of dawsonite, a mineral generally considered to be an important trap have been observed. These results question the importance of mineral trapping. At Springerville-St. Johns, CO2 occurs primarily as dissolved carbonate species and gas accumulations.

In order to understand the changes suggested by geologic evidence, we have modeled the hydrology of the system, including the reactive chemistry of the rock-fluid interactions on a broad scale. A TOUGH2/ChemTOUGH2 integrated finite difference model (see White et al., 2004). In this modeling we assume that the CO_2 in the reservoir results from a magmatic pulse of CO_2 and water lasting 5000 years beginning 10,000 years BP. This time fame provides flexibility for varying the pulse length and rock properties. Although the pulse was initiated 10000 years BP, we find that the modeled results and conclusions do not change if the pulse is initiated at a later time. In the simulation, the groundwater table is elevated as a result the pressure increases from the magmatic pulse. For simplicity, the effects of erosion by the Little Colorado River have been ignored. The cross-section is divided into elements with the element geometry determined by the need to reproduce interfaces between the geological layers (Figs. 3 and 4).

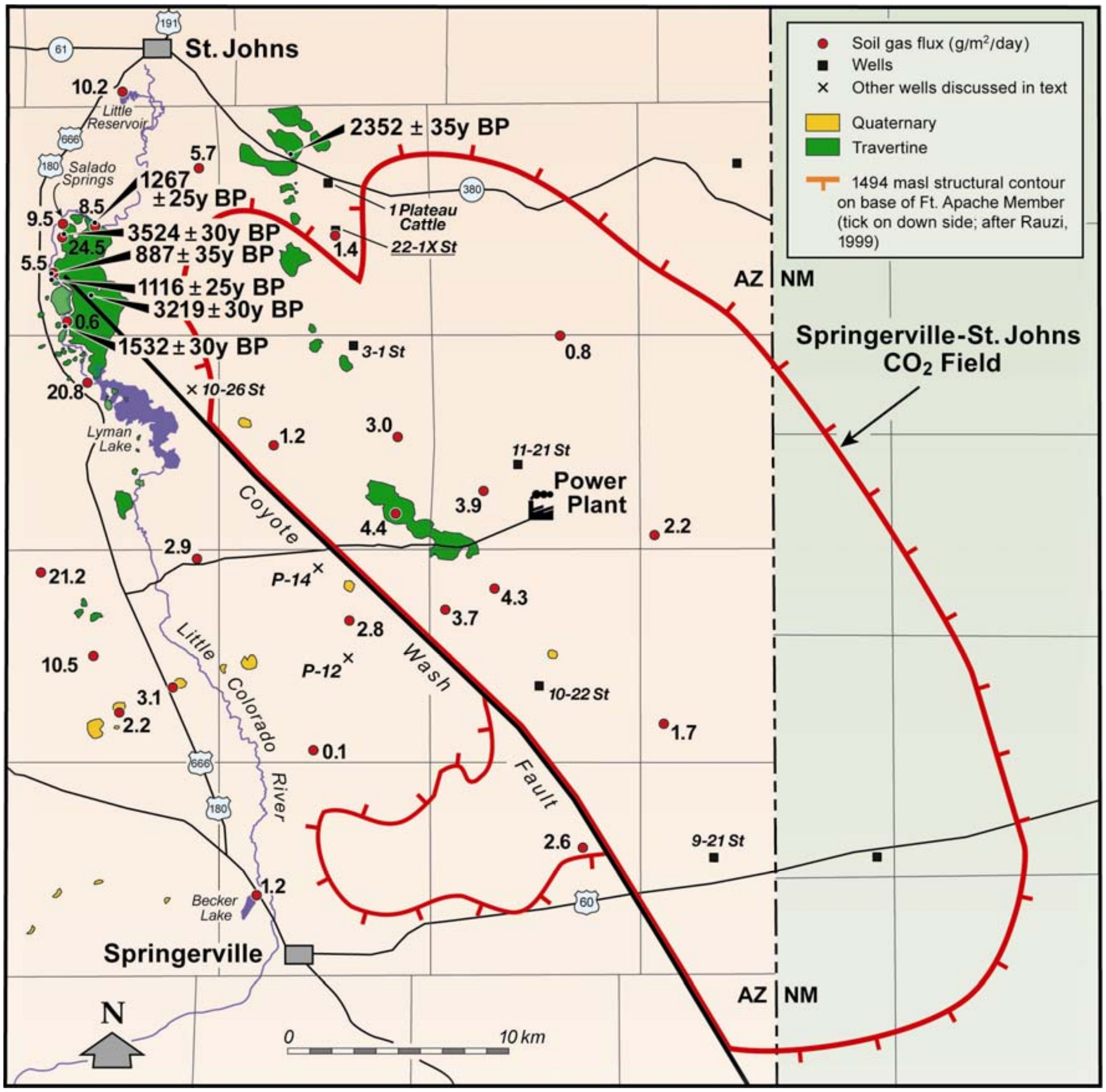


Fig. 2. Map of the Springerville-St. Johns CO_2 field area showing the locations of the travertine deposits, their ages in years BP (y BP), wells locations, and the results of soil gas flux measurements.

Table 1. ^{14}C AMS ages, locations and elevations of the travertine deposits at Springerville-St. Johns.

Sample #	mE	mN	Elevation (ft)	Age (y BP)	uncertainty
1	647247	3810848	5955	3524	30
2	646712	3808968	5880	887	35
3	662651	3798459	6970	undatable	
4	648682	3811171	5870	1267	25
5	647386	3806761	5900	1532	30
6	648539	3808007	6159	3219	30
7	657506	3814477	6240	2352	35
8	646838	3808876	5860	1116	25
Salado Spg. Outflow			5840	0	

1. Travertine dome near Salado Spgs; sample 1 m below rim, in vent
2. East side of dome, half way from summit; near Little Colorado River
3. Voight's Mesa; from travertine slab 100 m from central crater, west side of summit.
4. Upper flank of travertine slab on north-facing slope above Little Colorado River.
5. Dome, 10 m height on flood plain adjacent to Little Colorado River; sample from flank.
6. Dome on alluvial bench above sample 5; sample 30 cm below rim, inside crater.
7. Older vent area either side of road east of St Johns. Sample from northern side.
8. Small dome adjacent to Little Colorado River, flowed historically from vent (notch cut in side).

Boundary Conditions

Boundary conditions for the numerical model are

- Atmospheric pressure at the surface
- 0.3 cm annual infiltration
- Constant hydrostatic pressure on both the vertical boundaries
- No fluid flow over most of the base of the model
- A magmatic pulse of CO_2 (2 kg/s) and water (6 kg/s) flows into the base of the model for a period of 5000 years.
- Isothermal reservoir at 40^{o}C throughout.

Capillary pressure functions

Capillary pressure functions for the seal rocks within the Colorado Plateau are presented by White et al. (2004).

Permeability

The Permian Supai Formation consists of four members, the Corduroy Member, Fort Apache Member, Big A Butte Member and Amos Wash Member. There are multiple impermeable anhydrite and mudstone layers within these formations leading to vertical segregation of the CO_2 into several zones. It is not possible to represent all the anhydrite layers in the numerical model; however the two largest regions of anhydrite are included as low permeability layers

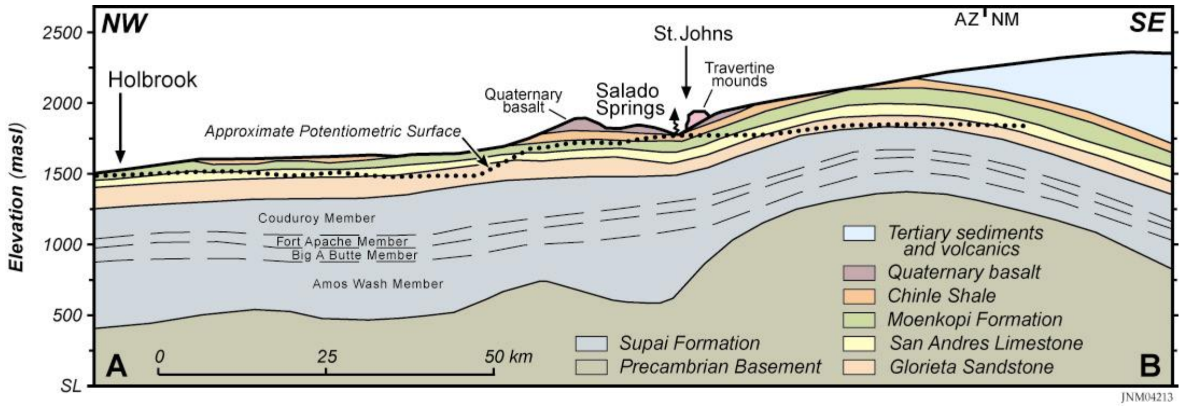


Fig. 3. Northwest-southeast cross-section of the Springerville-St Johns area. The CO_2 reservoir is confined to the Supai Formation within the structural high on the southeast end of the cross-section.

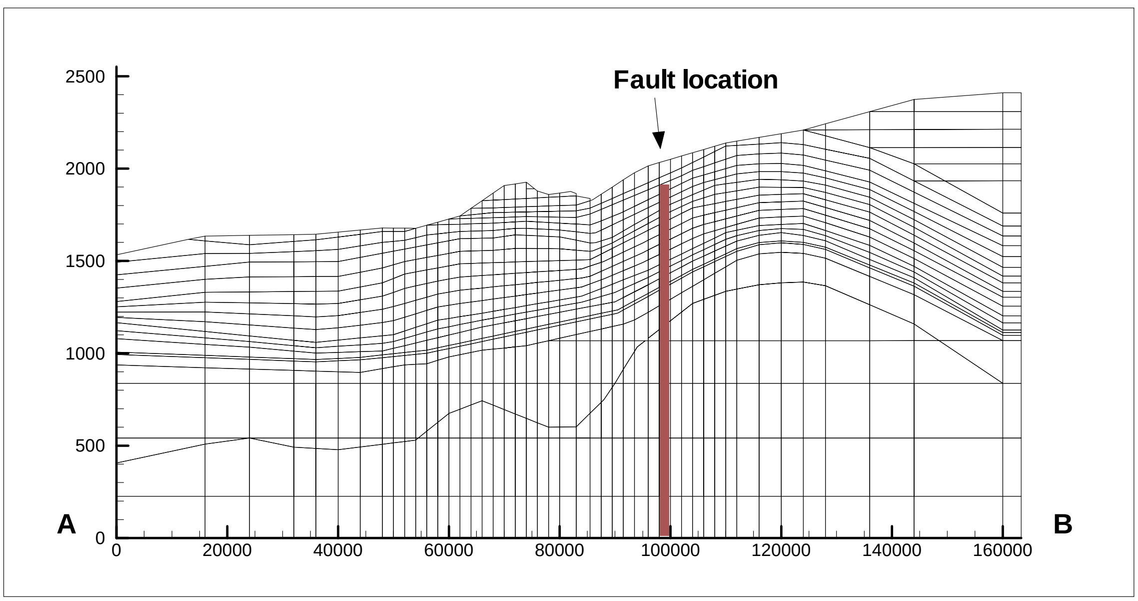


Fig. 4. Integrated finite difference grid used in modeling. The wavy lines are the interfaces between geologic units based on the cross section in Fig.3. Horizontal and vertical axes are in meters. The dense fault in the center of the section allows for a high permeability flow path.

in the Fort Apache and Big A Butte Members. There is a major fault in the vicinity of the CO_2 field that is represented in the model as a vertical region of high permeability cutting through the horizontal rock units (Fig. 4). Some model runs were attempted without the inclusion of this fault but it was found to be impossible to reproduce the present day distribution of CO_2 without including this fault. Parameters adopted for permeability and porosity are given in Table 2. These parameters are based largely on earlier modeling work at Farnham Dome (White et al. 2003, 2004)

Table 2: Rock properties used in all simulations.

Formation	Permeability (m^2)	Porosity
Basement	2.0×10^{-17}	0.02
Amos Wash	6.2×10^{-16}	0.10
Big A Butte	3.5×10^{-15}	0.17
Anhydrite	1.0×10^{-17}	0.05
Fort Apache	8.0×10^{-13}	0.16
Corduory	2.0×10^{-15}	0.16
Glorieta Sandstone	2.0×10^{-13}	0.10
San Andreas Limestone	2.0×10^{-13}	0.10
Moenkopi	2.0×10^{-17}	0.05
Chinle	2.0×10^{-17}	0.05
Volcanics	2.0×10^{-13}	0.10
Basalt	2.0×10^{-15}	0.10
Fault	1.0×10^{-12}	0.30

Mineralogy

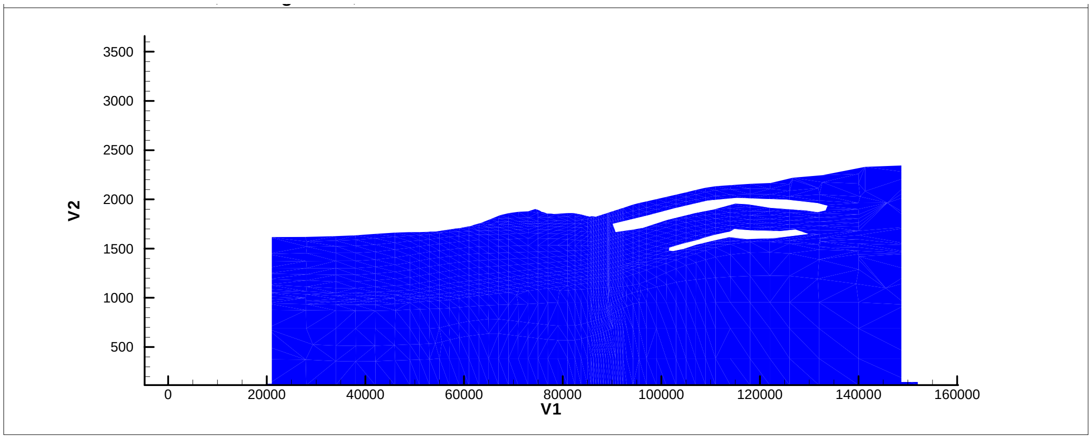
We have included sufficient geochemical complexity to represent the interaction of a simplified reservoir mineralogy (Table 3) with CO_2-rich water. Initial chemical conditions were calculated by first assigning the mineralogy specified in Table 3 to model elements, setting the reservoir fluid to a 0.03 M NaCl water, and then allowing the water to react with the reservoir for 10,000 years. This modified the original mineralogy slightly and provided initial conditions for the fluid reservoir throughout the reservoir.

Simulation Scenarios

The formation of the travertine was simulated assuming that a 5000 year pulse of CO_2 and water was added at the base of the model beneath the location of the CO_2 reservoir. This model was then run for about 10,000 years. CO_2 reservoir locations, gas saturations and mass fraction of CO_2 in the liquid phase are shown in Figure 5. The gas accumulations occur primarily in the Fort Apache and Big A Butte Members. These deep reservoirs are capped by layers of anhydrite; a third shallow reservoir is capped by the Moenkopi and Chinle Formations. Flows to the surface are small over most of the model with the exception of

Table 3. Mineralogy of the rock units used in the simulations.

Estimated mineralogy of Springerville-St Johns section (vol %)															
Formation/Unit	Porosity	Quartz	K-Feldspar	Na Feldspar	Ca Feldspar	Calcite	Dolomite	Gypsum	Na-smectite	Kaolinite	Illite	Hematite	Magnetite	Siderite	Pyrite	TOTAL
Tertiary Volcanics	20															
Chinle	5	45	1	1	1	2	2		20	20	1	1	1			100
Moenkopi	5	43	1	1	1	2	5		20	20		2				100
San Andres lst	10	19				2	65		2	2						100
Glorieta sst	20	71	1	1	1	2			2	2						100
Supai-Corduroy	12	63	4			5	5	15	3	3		2				100
Supai-Fort Apache	10	15	1	1		5	45	30	2	1						100
Supai-Big A Butte	17	63	2	2		5	5	15	3	3		2				100
Supai-Amos Wash	6	63	10	10	2	2			3	3		2	5			100
Precambrian	2	50	10	20	5	4			1	1	1		5		1	100



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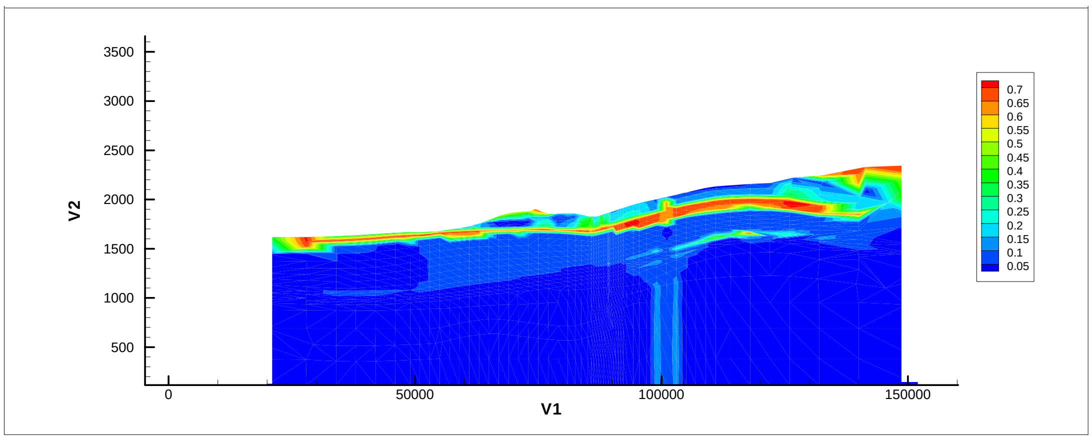

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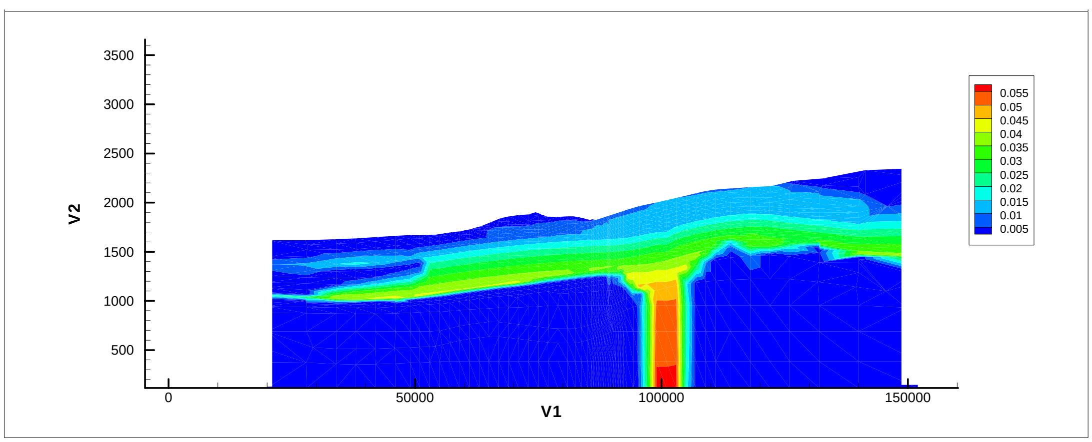

C

Fig. 5. Results of simulations 10,000 years after initiation of a CO_2 pulse. A. Location of gas reservoirs. B. Gas saturations. C. Mass fraction of CO_2 in the liquid phase.

significant flow from the fault zone associated with the travertine mounds during a 4000 year period, consistent with the age determinations.

Figure 6 shows the magnitude of the surface flow at several different times. 1000 years after the beginning of the pulse of CO_2, surface flows are at almost their peak values. The flows continue at this rate until the magmatic pulse ceases at 5000 years. Note that this figure shows only the flux of transported CO_2 dissolved in water flowing to the surface; there is also a significant flux of CO_2 in the gas phase, as can be seen in Figure 7. These surface flows are consistent with the observed travertine deposits. For example, the deposits near Lyman Lake and Salado Springs cover an area approximately 4 km wide (refer to Fig. 2). If we take a total water flow to the surface of 3.5×10^{-5} (from Fig. 7), from the reactive chemical transport simulation this water has a Ca^{2+} concentration of 0.0165 M. Assuming that all the calcium is deposited as travertine, that the travertine has a density of 1300 kg/m^3 and that 30% of the surface area is covered by travertine, we arrive at a travertine thickness of 6.1 meters deposited over the 4000 year period of high surface flows.

In summary, the models indicate that a pulse of CO_2 and water lasting 5000 years is needed to produce realistic flow and travertine deposition rates. Although our model was run for 10,000 years, with the pulse being initiated at 5000 y BP, identical results are obtained if the pulse is initiated at 5000-6000 years BP. Initiating the pulse at a later time, will produce travertine deposits less than 4000 years old.

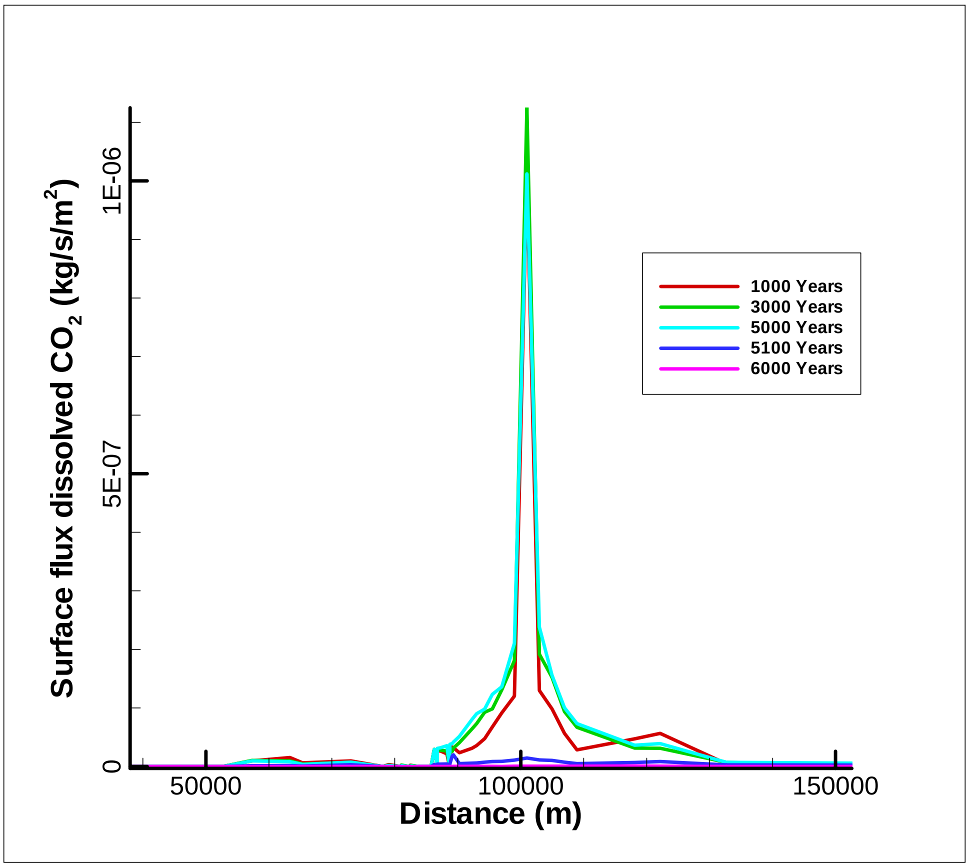

Fig. 6: Surface flux of dissolved CO_2 at Springerville-St. Johns at different times after initiation of the CO_2 pulse.

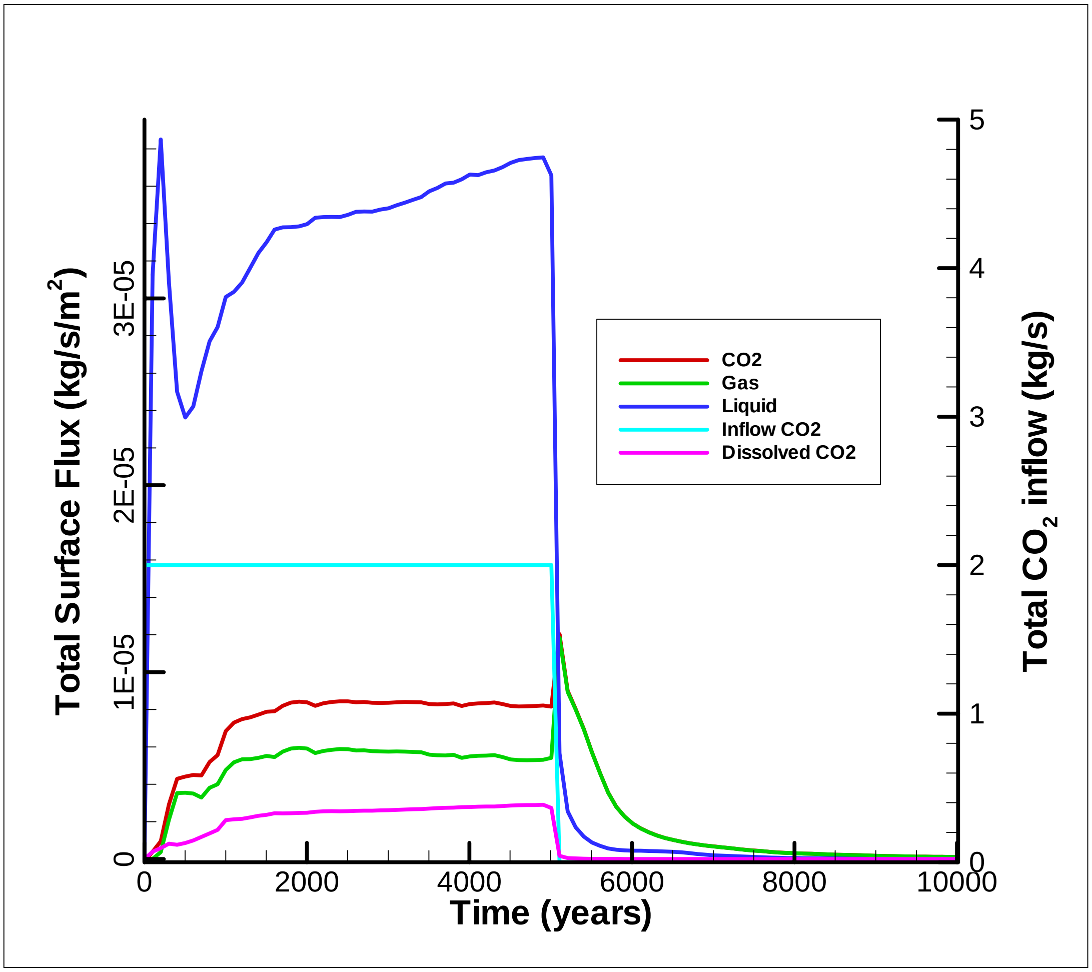

Fig. 7. Total flows to the surface at Springerville-St. Johns as a function of time. In the model, time 0 occurs at 10,000 years BP.

<u>CO_2 Flux Measurements</u>

Soil CO_2 flux surveys have been carried out over three regions with natural occurrences of CO_2 on the Colorado Plateau (Allis et al., 2005; White et al., 2004). At Farnham Dome, Utah, and Springerville-St. Johns, proven CO_2 reservoirs occur at 600 - 800 m depth, but no anomalous CO_2 flux was detected. Background fluxes of up to about 5 g m^{-2} day^{-1} were common in poorly vegetated, arid areas, and up to about 20 g m^{-2} day^{-1} were found at Springerville-St. Johns in heavily vegetated, wet ground adjacent to springs. The higher fluxes are attributed to shallow root zone activity rather than to a deep upflow of CO_2. At the Crystal Geyser-Ten Mile Graben in Utah (Figs. 8-10), localized areas of anomalously high CO_2 flux (~ 100 g m^{-2} day^{-1}) occur along a fault zone near visibly degassing features. Isotopic measurements on CO_2 collected from nearby springs indicate that it originated at depth. Evidence of widespread vein calcite at the surface (Farnham Dome) and travertine deposits at the other two areas suggests that discharge of CO_2-rich fluids has occurred in the past. Despite the lack of evidence for significant present day leakage of CO_2 to the atmosphere at Springerville-St. Johns and Crystal Geyser-Ten Mile Graben, there are

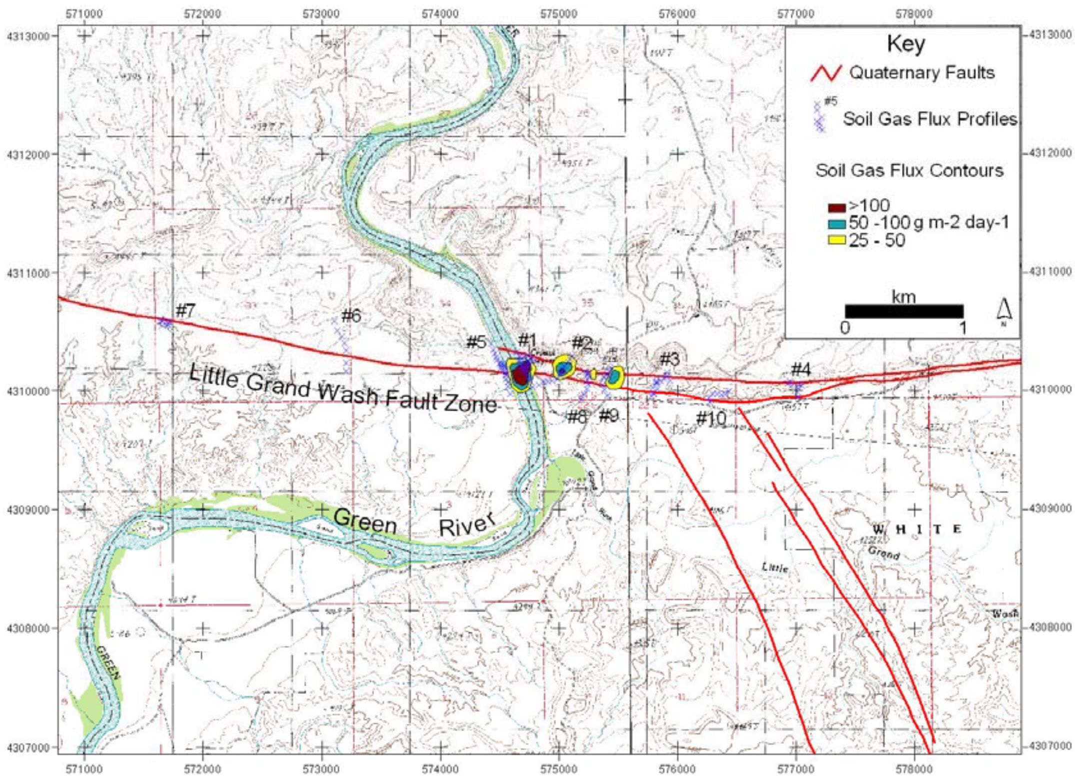

Figure 8. Location of 10 soil CO_2 flux profiles along the Little Grand Wash Fault zone at Crystal Geysers, UT. Fault locations mapped by Doelling (2001). Each cross represents a single flux measurement. Green areas show vegetation.

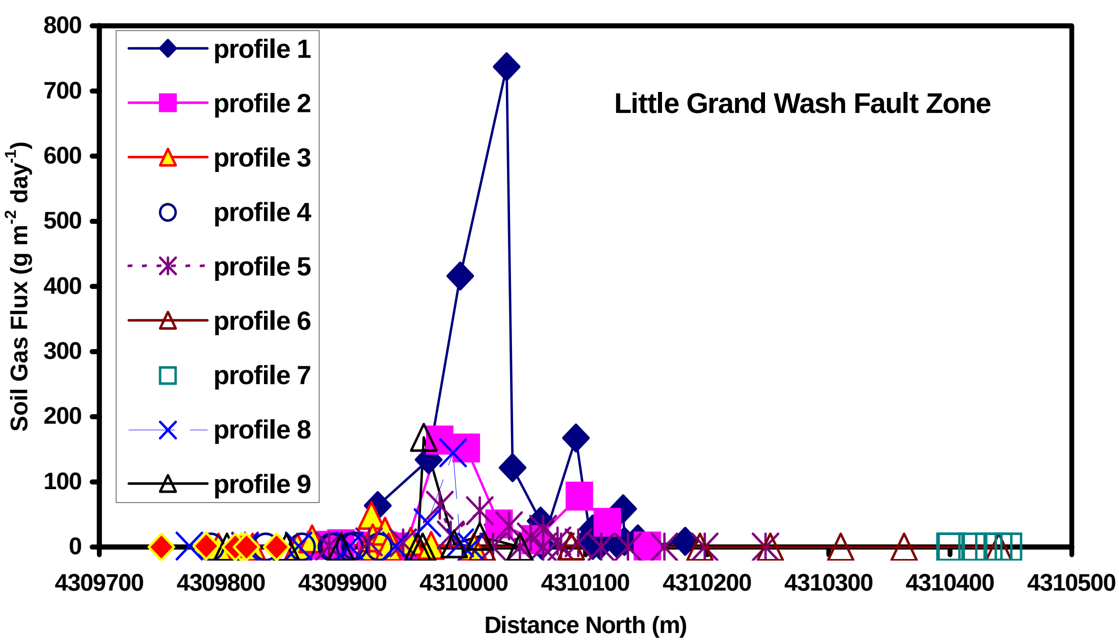

Fig. 9. Pattern of CO_2 flux results from Crystal Geysers obtained by superimposing data based on the northing coordinate of the measurement site.

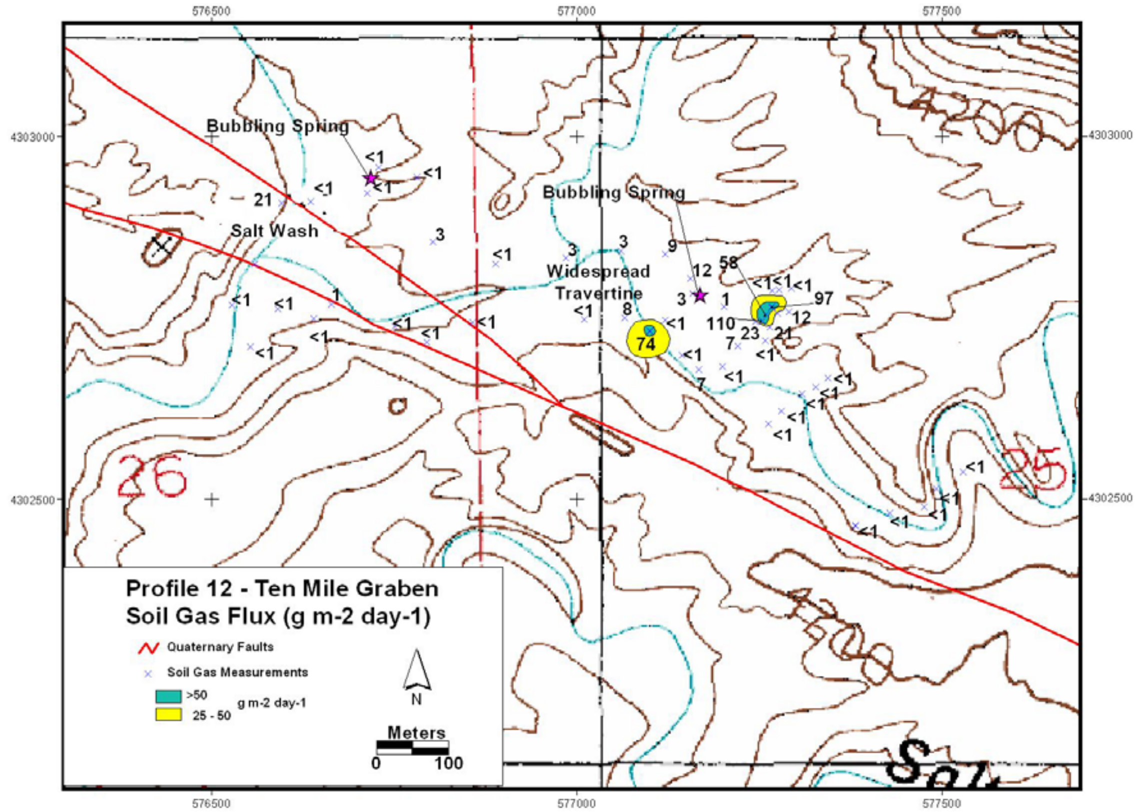

Fig. 10. Details of CO_2 flux results at the west end of Ten Mile Graben (Profile 12). Faults are from Doelling (2001).

significant outflows of high-bicarbonate water in both areas suggesting continuous migration of CO_2 in the aqueous phase from depth.

Numerical Simulations

Numerical simulations were conducted using the reactive chemical transport code ChemTOUGH2 (White, 1995) to examine the mobility of CO_2 within non domed geologic structures that do not provide the conditions required for the development of artificial CO_2 reservoirs. The purpose of this investigation was to determine if broad asymmetrical anticlines, such as the San Rafael Swell, could serve as sites for CO_2 sequestration. The San Rafael Swell is one of numerous uplifts on the Colorado Plateau. It was chosen as an example because it is collocated with the Hunter power plant. The premise of this study is that very long flow paths between the injection point and surface will allow for the permanent sequestration of CO_2 as a mineral or as a dissolved constituent in the ground water. The full text of the manuscript is presented below in Appendix 2.

CONCLUSIONS

Few investigations of naturally occurring CO_2 reservoirs have been conducted with the goal of evaluating the factors that will influence the mobility and behavior of CO_2 in

1. Investigations of natural CO_2 reservoirs have provided information on the mobility of CO_2 that was not anticipated.

2. Leakage of CO_2 cannot be predicted. At Springerville-St. Johns and Farhnam Dome, no surface flux of CO_2 was detected, despite the presence of shallow CO_2 reservoirs.

3. Despite the lack of evidence for significant present day leakage of CO_2 to the atmosphere at Springerville-St. Johns there are significant outflows of high-bicarbonate water suggesting continuous migration of CO_2 from depth. Disssolved CO_2 could be the main source of CO_2 leakage from a sequestration site. Chemical analyses of ground water must be included in any monitoring program.

4. Leakage of CO_2 may occur episodically. At Springerville-St. Johns, outflows of ground water containing dissolved CO_2 deposited extensive sheets and domes of travertine (calcite) during the last 4000 years. CO_2 not trapped as calcite was discharged to the atmosphere.

5. At Crystal Geyser-Ten Mile Graben, leakage occurs at isolated points along fault zones. The very localized nature of the CO_2 flux anomalies, and presents challenges for effective, long term monitoring of CO_2 leakage.

6. At Springerville-St. Johns, the permanent trapping of CO_2 in mineral phases appears to be insignificant. Only trace amounts of dawsonite, a mineral generally considered to be an important trap have been observed. The bulk of the CO_2 occurs as gas accumulations and dissolved carbonate species in the ground water.

7. Broad asymmetrical anticlines occurring beneath the Colorado Plateau have the potential to sequester CO_2 from collocated power plants. The mobility of CO_2 after 30 years of injection was numerically simulated. At 1000 years, 21% of the CO_2 was found to be permanently sequestered in mineral phases, 52% occurred as a gas or was dissolved in ground water and 17% had leaked to the surface. Leakage stopped after 1500 years, at which point 70% of the CO_2 was permanently sequestered.

8. The importance of mineral trapping may be overestimated. Further investigations of natural systems are required to assess its importance.

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